Multi-Scale Monitoring and Prediction of System Responses to Biostimulation

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I. INTRODUCTION

To advance solutions needed for remediation of DOE contaminated sites, approaches are needed that can elucidate and predict reactions associated with coupled biological, geochemical, and hydrological processes over a variety of spatial scales and in heterogeneous environments. Our laboratory experimental experiments, which were conducted under controlled conditions, suggest that geophysical methods have the potential for elucidating system transformations that often occur during remediation, such as the generation of gases and precipitates. In this new ERSP project, we will Integrate hydrological, biogeochemical, and geophysical expertise and approaches to:

- Explore the potential of geophysical methods for detecting changes in physical, chemical, and biological properties at the field scale
- Explore the joint use of reactive transport modeling and geophysical monitoring information for improvements in both methods.

A brief review of our previously-conducted laboratory results are given in Section II. Section III Describes the approach for our new project, which will have both laboratory and field-scale components. The field scale component will be conducted at the Rifle, CO. site, which is described in Section IV.

II. PREVIOUS EXPERIMENTAL RESULTS

With EMSP support, we have recently investigated the utility of the following different geophysical techniques for detecting various system transformations at the column scale:

–Radar velocity measurements for measuring the evolution of gas during denitrification. (Hubbard and Williams, 2004).

–Seismic measurements to detect onset of gas evolution during denitrification

–Self Potential (SP) measurements for characterization of redox conditions. We have used the SP method to track the onset and location of microbial sulfate-reduction in saturated sediments at the laboratory scale during conditions of organic carbon amendment. The experimental results suggest the ability to measure the changes in the spatiotemporal location of sulfate-reduction during bioremediation (Williams et al., 2005b)

–Seismic Amplitude and Induced Polarization (IP) methods to track the formation and aggregation of precipitates associated with sulfate reduction

–IP methods to track changes in iron mineralogy during remediation. We have used IP methods to track physiochemical changes in iron-bearing clays resulting from microbial respiration, which correlated with the exhaustion of bioavailable ferric compounds. This study shows that IP methods may be a reasonable approach for noninvasively monitoring the sustainability of prolonged iron-reduction under stimulated conditions (Williams et al., 2005a; See Section IV).

–Combined seismic and IP methods to monitor biomass accumulation. Seismic and IP methods were used to characterize the accumulation of biomass within saturated sediments. The experimental conditions were similar to those used during our studies of metal sulfide precipitation, with the exception that no metals were added to the influent solution. Although destructive evaluation of the sediments revealed significant biofilm development, only modest changes in IP and seismic signatures were observed (Ntarlagiannis et al., 2005).

Examples of a few of these results are briefly described below.

Monitoring Gas Evolution during Denitrification

Laboratory-scale biostimulation experiments were performed to assess the seismic and radar responses to gas generation using instrumented columns such as those shown in Figure 1. Using a three phase mixing model (gas, water, solids) with radar velocity measurements, the volume of pore space filled with evolved N2 gas was estimated to be within 1% of that obtained using gravimetric measurements (Figure 2; Hubbard and Williams, 2004). Seismic methods indicated that the presence of gas in the pore space dramatically attenuates the signal (Figure 3; Williams 2003).

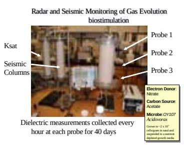
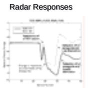
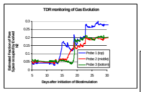


Figure 1

Radar Responses





Investigation Approach:	Value at beginning of experiment	Value at end of experiment	Interpretation of time-lapse changes
Dielectric from Radar Velocity	24.8 (average)	19 (average)	24.6 % of pore space occupied by evolved N_2 gas
Column Weight	7027 g	6718 g	23.3% of pore space occupied by evolved N_2 gas.
Radar Amplitude	Low	High	Evolved gas in pore space attenuates signal less than water in pore space
Hydraulic Conductivity	0.0038 cm/s	0.0017 cm/s	55% reduction in hydraulic conductivity

Figure 2

Results of Seismic Monitoring: Seismic Amplitude decreased as N_2 increased

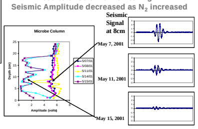


Figure 3

Monitoring of Sulfide Production

Column scale studies (Figure 4) were performed to assess the influence of evolved aqueous sulfides as well as the development of precipitates on geophysical responses. After poising the system to undergo sulfate reduction, *Desulfovibrio vulgaris* were added to the middle of the column and the seismic and complex electrical responses were recorded. These studies showed how both seismic amplitude (Figure 5) and Induced Polarization (Figure 6) corresponded to the initiation and aggregation of precipitates over space and time (Williams et al., 2005a; Ntarlagiannis et al., 2005).

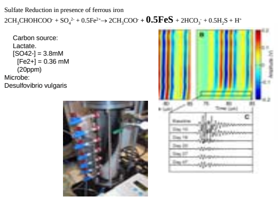


Figure 4

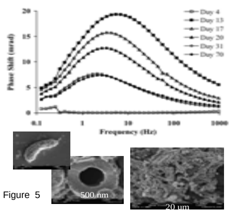


Figure 5

ACKNOWLDGEMENTS

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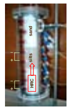
III. NEW STUDY: RESEARCH DESIGN AND METHODS

Our proposed research includes theoretical, numerical, and experimental investigations performed at the laboratory and the field scale, which involve the remote monitoring and prediction of biogeochemical processes using geophysical methods and reactive transport modeling, respectively. Linkage between the laboratory-scale and field-scale investigations will be ensured by using the same (native) sediments (and in some cases, groundwater), and by using the same geophysical measurement techniques and reactive transport code at both scales. By investigating the geophysical response to coupled processes and by performing calibrated and validated reactive transport modeling at both scales under the same environmental conditions, we can begin to investigate how novel monitoring and modeling approaches scale with space and time.

KEY COMPONENTS

Laboratory Experiments

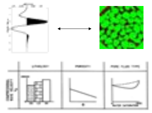


Laboratory studies will performed to mimic conditions at the Old Rifle, CO, field site (see Section IV) using native groundwater and sediments. Radar, seismic, SP, and complex resistivity measurements will be conducted during the experiment concomitant with geochemical, hydrological, and microbiological measurements. Specific experiments that will be conducted include the Geophysical Monitoring of:

- Ferric Oxide Reduction during Biostimulation;
- Stimulation of Single Microbial Strains in Native Sediments;
- Stimulation of Mixed Microbial Strain in Native Sediments;
- FeS Oxidation.

Through these experiments, we will determine if FeRB and SRB activity and their impact on aquifer sediments can be uniquely detected via geophysical methods.

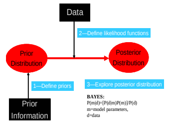
Petrophysical Relationships



The experiments illustrated in Section II showed that correlations exist between the changes in geophysical responses and the changes in biogeochemical properties. To date, however, we have interpreted these changes only qualitatively, based on comparing the changes in geophysical signatures with biogeochemical measurements. In this component of the project, we will theoretically investigate the controls of such biogeochemical products on complex electrical and seismic amplitude responses in saturated, porous media and test the models using the laboratory data.

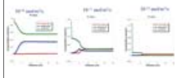
We anticipate that this modeling will enable a more quantitative interpretation of the geophysical signatures than has been heretofore possible

Estimation Framework



Both the laboratory experiments and the field-scale experiments (described below) will result in system transformations that will yield multiple products (such as gases, precipitates, and biofilms) within a heterogeneous environment. In this component, ***we will develop a stochastic framework that can be used with petrophysical models and measured data to estimate the temporal and spatial distribution of system transformations.*** We will consider the biogeochemical parameters and geophysical attributes as spatial and temporal random variables, and will explore the use of the biogeochemical, hydrogeological, and geophysical parameters as continuous values, indicators, or categorical values. We will use the Markov Chain Monte Carlo approach for integrating the disparate datasets and relationships.

Reactive Transport Modeling



This component will focus on validating and/or calibrating reactive transport models against carefully controlled laboratory experiments. We will use the pore-water chemistry determined over the course of the experiment and the solid-phase mineralogy determined via post mortem analysis to develop a defensible description of the reaction network (pathways and rates). The reactive transport modeling will be carried out with the CRUNCH (Steefel et al., 2003) and TOUGHREACT codes. The reactive transport model will be calibrated to the geophysical data, but only by using the independent constraints provided by the microbiological, chemical, and physical data. This is a key step, since the geophysical data will be crucial in developing a high-resolution data set at the field scale, where complete microbiological, chemical, and physical characterization of the subsurface material will not be feasible.

IV. RECENT RESULTS: FIELD and LABORATORY INVESTIGATIONS ASSOCIATED WITH THE RIFLE SITE

Ongoing work within the alluvial aquifer at Rifle, CO, has focused on investigating the utility of electron-donor amendments for facilitating microbial reduction of U(VI) to U(IV) (Long et al., 2005; Vrionis et al, 2005) through a series of experiments in different flow cells conducted from 2002-2005. Early experiments at the site showed that U(VI) loss from groundwater occurred synchronously with growth of Geobacter after acetate amendment, and illustrated the importance of maintaining iron-reducing conditions for optimal U(VI) removal. Since the interplay between iron and sulfate reduction is believed to be of critical importance to the processes ongoing at this site, quantitative interpretation of geophysical data in terms of redox state, exhaustion of bioavailable iron mineral phases, or onset of sulfate reduction should benefit the understanding and sustained remediation of uranium at the site.

Complex surface resistivity data were collected before and after stimulation at two flow cells within the Rifle Study Area (Figures 6 and 7) to measure both electrical conductivity and phase. Changes in phase result from a change in the ability of the material to conduct a current. Figure 8 shows phase difference cross sections within two different flow cells a few weeks after stimulation and at ten weeks after stimulation. These images show that the phase response is quite different within the two cells. In the 2004 test cell, the response is interpreted to be associated with a change in clay mineralogy associated with iron reduction, whereas the phase response associated with the 2005 experiment is indicative of the formation of FeS precipitates. Crosshole electrical data collected during the 2005 experiment also indicated that much of the electrical response is confined to a higher hydraulic conductivity layer.

Through field characterization and monitoring using seismic, radar, complex electrical and SP measurements, and through coupling the field characterization with the lab-scale experiments and reactive transport modeling described above, we will investigate at the Rifle Site:

- **Can geophysical data be used to distinguish between iron and sulfate reduction processes and track these process over space and time?**
- **What is the role of heterogeneity on the system transformations?**

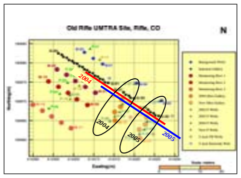


Figure 6

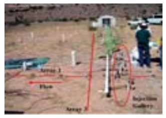


Figure 7

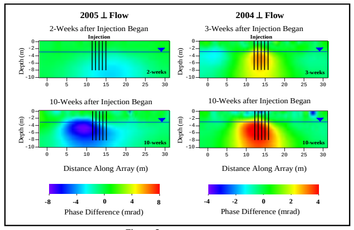


Figure 8

To address some of these questions, recent **LABORATORY EXPERIMENTS** have examined the change in the complex resistivity response of a model phyllosilicate (e.g. nontronite) following iron- and sulfate-reduction. Figure 9 illustrates the change in the phase response of an initially oxidized nontronite-sand mixture (0.8% w/w) following stimulated microbial alteration, with the iron- and sulfate-reducing strains isolated from the Rifle clay-sized fraction. In agreement with our 2004 field observations, the magnitude of the phase response ***decreased*** following iron-reduction, most likely the result of decreasing particle surface area following reordering of the clay particles (Figure 10). In contrast, exposure of the clays to H_2S following sulfate-reduction led to blackening of the material (e.g. FeS) and an ***increase*** in the phase response (Figure 5). Such a result is in agreement with our 2005 field observations, where sulfate-reduction was found to be an important terminal electron accepting pathway. Ongoing work with native Rifle sediments is being performed to test similar hypotheses.

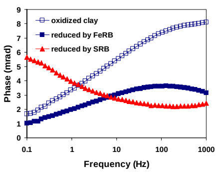


Figure 9

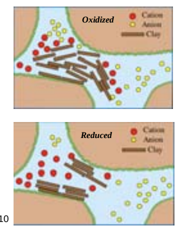


Figure 10

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